

SULFUR-TOLERANT CATALYST SYSTEMS**STATEMENT REGARDING FEDERALLY
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BACKGROUND

The performance of catalyst systems, particularly those that are nickel-based, can be significantly degraded by sulfur, and/or sulfur-containing compounds, which poison the catalyst. For instance, hydrogen sulfide, which is present in most common fossil fuel sources, can cause a significant poisoning of nickel-based anodes in solid oxide fuel cells (SOFC). In a specific example, a nickel-YSZ-based anode can typically show a 35-50% drop in power within 20 hours when even one ppm of hydrogen sulfide is present in the fuel. While desulfurizers can be employed to treat sulfur-containing reactants prior to reacting them on a nickel-based catalyst, such desulfurizers are often costly and/or bulky. Accordingly, a need exists for nickel-based catalyst systems having improved sulfur tolerance.

SUMMARY

Aspects of the present invention are encompassed by methods for improving sulfur tolerance in catalyst systems and by the improved catalyst systems themselves. Specifically, according to some embodiments, methods for improving sulfur tolerance in catalyst systems comprise adding praseodymium to the catalyst system, thereby inhibiting sulfur poisoning of the catalyst system. Suitable catalyst systems can include those that are susceptible to sulfur poisoning, and can include, but are not limited to those based on Ni, Rh, and Pt. In other embodiments, methods can further comprise adding a combination of either ruthenium and praseodymium or cerium and praseodymium to the catalyst system. A catalyst system, as used herein, comprises a catalyst having metal, metal alloys, and/or metal-cermet for standard catalysis applications (e.g., reformation, etc.) or for alternative applications (e.g., electrochemistry on fuel cell electrodes). Exemplary metals can include, but are not limited to Ni, Rh, and Pt.

In specific embodiments, the adding of praseodymium, or of either praseodymium and ruthenium or praseodymium and cerium, to the catalyst system can comprise infiltrating the catalyst system with a solution comprising the praseodymium, the praseodymium and ruthenium, or the praseodymium and cerium. Alternatively, the adding can comprise the addition of powders comprising praseodymium, praseodymium and ruthenium, or praseodymium and cerium to a feedstock for the catalyst system and the subsequent application of powder processing techniques. The methods described herein of adding Pr-based materials are preferred embodiments and are not intended to preclude from the scope of the present invention the use of other methods that may produce equivalent results.

In a particular embodiment, the method of adding praseodymium alone, or in combination with Ru and/or Ce, is performed in a solid oxide fuel cell (SOFC), wherein a fuel-cell anode comprises a nickel-based catalyst system. In one such embodiment, the nickel-based catalyst system can comprise porous nickel-YSZ.

In other embodiments, the present invention can also encompass a catalyst system comprising an added amount of praseodymium sufficient to inhibit poisoning of the system by sulfur. The praseodymium can exist in mixed valence states.

For example, the catalyst system can comprise a mixed phase of Pr^{3+} and Pr^{4+} . Furthermore, the catalyst system can also comprise ruthenium and/or cerium. In a specific embodiment, a nickel-based catalyst system comprises nickel-YSZ, and is part of an anode in a fuel-cell.

The purpose of the foregoing abstract is to enable the United States Patent and Trademark Office and the public generally, especially the scientists, engineers, and practitioners in the art who are not familiar with patent or legal terms or phraseology, to determine quickly from a cursory inspection the nature and essence of the technical disclosure of the application. The abstract is neither intended to define the invention of the application, which is measured by the claims, nor is it intended to be limiting as to the scope of the invention in any way.

Various advantages and novel features of the present invention are described herein and will become further readily apparent to those skilled in this art from the following detailed description. In the preceding and following descriptions, various embodiments, including the preferred embodiment, of the invention, are shown and described, at least, by way of illustration of the best mode contemplated for carrying out the invention. As will be realized, the invention is capable of modification in various respects without departing from the invention. Accordingly, the drawings and description of the preferred embodiment set forth hereafter are to be regarded as illustrative in nature, and not as restrictive.

DESCRIPTION OF DRAWINGS

Embodiments of the invention are described below with reference to the following accompanying drawings.

FIG. 1 is a bar graph showing the performance of various Ni-based catalyst systems, according to embodiments of the present invention, as implemented in a fuel cell structure.

FIG. 2 shows electrochemical performance of various cells after introduction of H_2S with 50/50 N_2/H_2 fuel.

DETAILED DESCRIPTION

The following description includes the preferred best mode of one embodiment of the present invention. It will be clear from this description of the invention that the invention is not limited to these illustrated embodiments but that the invention also includes a variety of modifications and embodiments thereto. Therefore the present description should be seen as illustrative and not limiting. While the invention is susceptible of various modifications and alternative constructions, it should be understood, that there is no intention to limit the invention to the specific form disclosed, but, on the contrary, the invention is to cover all modifications, alternative constructions, and equivalents falling within the spirit and scope of the invention as defined in the claims.

According to some embodiments of the present invention, praseodymium alone, or in combination with ruthenium and/or cerium, can be added to the nickel-based catalyst system through infiltration of the catalyst system with a solution comprising the praseodymium, praseodymium and ruthenium, or praseodymium and cerium. For example, in a particular instance, infiltration of an anode in a fuel cell by Pr-based materials was conducted using aqueous solutions of praseodymium (III) nitrate hydrate ($\text{Pr}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$), nickel (II) nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), cerium (IV)